

Optical pumping and non-destructive readout of a single magnetic impurity spin in an InAs/GaAs quantum dot

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Abstract

We report on the resonant optical pumping of the $|\pm 1\rangle$ spin states of a single Mn dopant in an InAs/GaAs quantum dot embedded itself in a charge tuneable device. The experiment relies on a “W” scheme of transitions reached when a suitable longitudinal magnetic field is applied. The optical pumping is achieved via the resonant excitation of the central Λ system at the neutral exciton X^0 energy. For a specific gate voltage, the red-shifted photoluminescence of the charged exciton X^- is observed, which allows non-destructive readout of the spin polarization. An arbitrary spin preparation in the $|+1\rangle$ or $|-1\rangle$ state characterized by a polarization near or above 50% is evidenced.

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The possibility to precisely prepare and control the quantum state of an individual spin in a quantum dot (QD) is an appealing issue both from a fundamental point of view and for potential applications in the field of quantum information [1]. Recent works have demonstrated that the spin of a an electron localized in a gate-defined or self-assembled semiconductor QD can be coherently manipulated [2–5]. However, this system is not really ideal because of its intrinsic coupling to the environment (phonons, nuclear spins or surrounding charges) which restrains its potential for future developments. QDs doped with a single magnetic impurity like a Mn atom offer an interesting alternative in this regards [6]. Such isolated magnetic spin which is provided by core electrons ($3d^5$ for Mn), is potentially better isolated from the environment than valence or conduction electrons. Besides, thanks to the *sp-d* exchange which yields spin-dependent sub-levels, selective addressing by optical methods is in principle attainable. A singly Mn-doped QD could reveal itself as particularly suitable for the realization of quantum bits.

Among recent investigations in this direction, the initialization of a Mn spin in II-VI semiconductor QDs has been demonstrated via optical pumping (OP) [7, 8]. In II-VI host materials, the $5/2$ Mn spin which presents 6 distinct states is however not ideal for quantum operations. In III-V QDs the Mn effective spin turns out to be much simpler : a Mn atom forms indeed an acceptor state denoted A^0 (a negative center A_{Mn}^- and a tightly bound hole) presenting a total angular momentum reduced to $J = 1$ in its ground state [9, 10]. Furthermore, in a non-spherical self-assembled QD, the state $|J_z = 0\rangle$ is substantially higher in energy than the two circular states $|J_z = \pm 1\rangle$ by a few meV [11, 12]. At low temperature these states $|\pm 1\rangle$ define a 2-level system, with whole occupation near unity, which we refer to as the A^0 spin in the following. Due to the large QD-induced anisotropy [12], A^0 spin initialization cannot be achieved following a simple non-resonant approach as used recently for the ionized impurities A_{Mn}^- in bulk GaAs [13, 14] and based on *sp-d* interaction with spin polarized electrons. An OP scheme dedicated to the case of A^0 spin in a self-assembled QD is required.

In this Letter, we present an OP experiment in which the A^0 spin in an InAs/GaAs QD is pumped arbitrarily into one of its $|\pm 1\rangle$ states by using a resonant laser excitation. Its principle relies on a “W” energy level scheme which arises at a specific longitudinal magnetic field, similarly as quantum dot molecules in an electric field [15, 16]. Besides, the spin initialization and its readout are spectrally dissociated by using a technique recently

introduced by Kloeffel *et al.* [17] which enables to monitor the spin population by using a standard micro-photoluminescence (μ -PL) setup. Efficient OP is demonstrated through the vanishing (resp. enhancement) of transitions associated to the pumped (resp. populated) A^0 level.

The sample used in this work consists of a Mn-doped QD layer embedded in between an electron reservoir and a Schottky gate allowing the control of the QD charge. The Mn doping is very dilute and less than 1% of the QDs are effectively doped by a single Mn impurity, see Ref. [11] for details. The μ -PL spectroscopy of such individual QDs is carried out with a 2 mm focal length aspheric lens (N.A. 0.5) actuated by piezo-stages and mounted in a split-coil magneto-optical cryostat. The collected PL is dispersed by a 0.6 m-focal length double spectrometer and detected by a Nitrogen-cooled CCD array camera providing a $\sim 20\mu\text{eV}$ spectral resolution. All measurements presented in this Letter were performed at low temperature ($T \sim 2$ K) with a magnetic field applied parallel to the optical axis (Fig. 1(a) insert).

The QD studied in this work contains a Mn impurity in the A^0 configuration [9, 10, 18]. This acceptor is characterized by an antiferromagnetic (AFM) p - d exchange between the $3d^5$ Mn spin $S = 5/2$ and the spin $J_h = 3/2$ of the bound hole which gives rise to 4 levels as a function of the total angular momentum $J = 1, 2, 3$ or 4 . At low temperature ($T \sim 2$ K) only the ground state $J = 1$ is significantly occupied so that higher states ($J = 2, 3, 4$) can be neglected. Moreover, the lens shape and biaxial strain of InAs self-assembled QDs shift the $|J_z = 0\rangle$ state of the $J = 1$ triplet to a higher energy by about 2 meV [12]. Consequently, this level is also negligibly occupied at 2 K and one can describe the A^0 impurity as an effective 2-level system associated to the spin projections $J_z = \pm 1$. A natural basis to describe the A^0 -QD states in presence of additional (photo-created) carriers in the QD S-shell reads $|\pm 1, J_z^{\text{QD}}\rangle$, where J_z^{QD} denotes the z -projection of their total spin.

Figure 1(a) shows for the investigated QD the typical signature of A^0 in PL spectroscopy as a function of a positive longitudinal magnetic field. For the gate voltage used here ($V_g = 0.6$ V) the QD is charged by an extra electron yielding under optical excitation a negative trion X^- (a hole (h) with 2 electrons (e^-)). Since both conduction e^- are paired in a singlet configuration in the QD S-shell, its spin is determined by the h spin only and hence reads $J_z^{\text{QD}} = \pm 3/2$ ($\uparrow\uparrow$ or $\downarrow\downarrow$). A detailed discussion of the spectral signature of Fig. 1(a), *i.e.* two intense lines denoted FM and AFM which anticross with two weaker lines forming

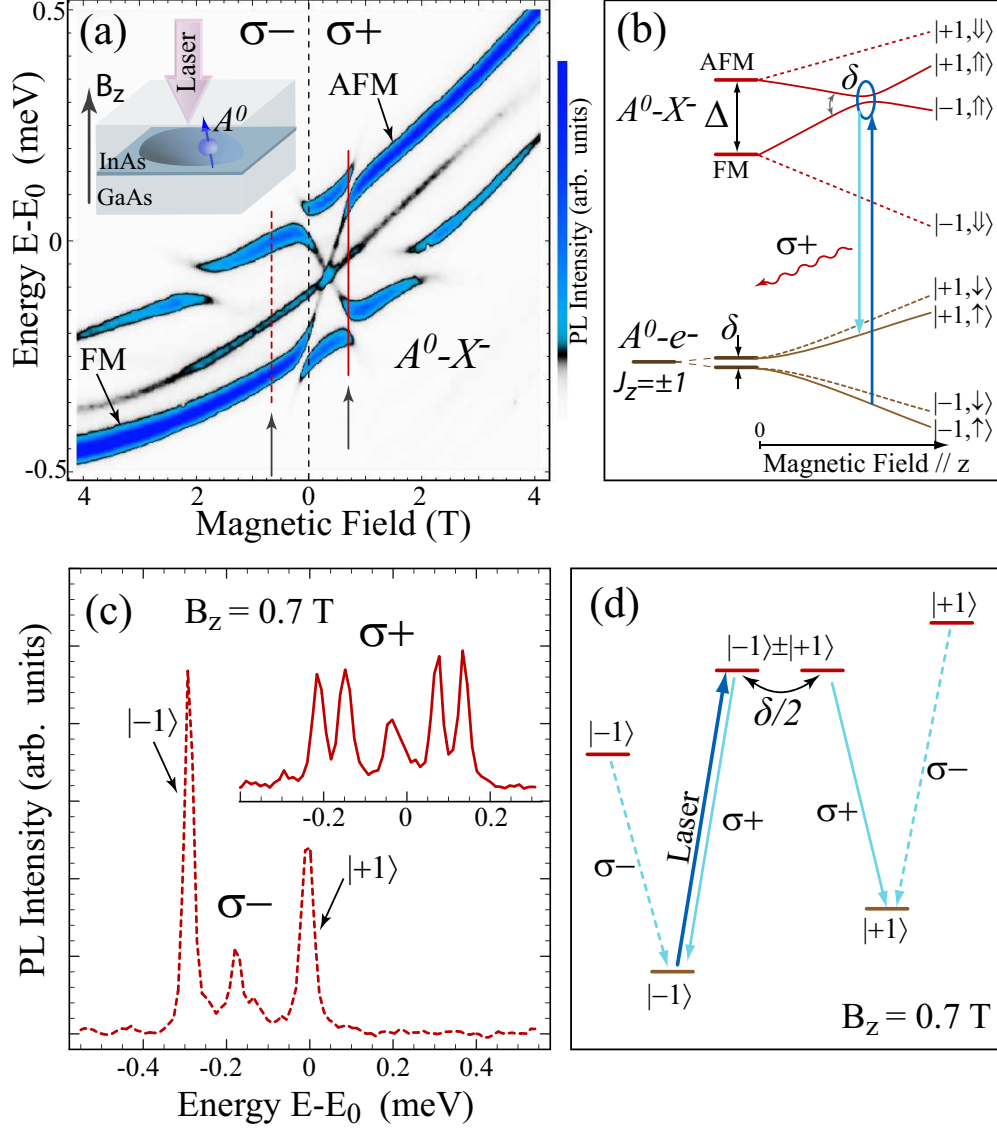


FIG. 1. (a) Density plot of μ -PL intensity under non-resonant excitation (HeNe at 633 nm) for a Mn-doped QD in magnetic field. Detection is σ^+ or σ^- polarized as indicated and centered around $E_0 = 1.274$ eV. (b) Level diagram corresponding to the main transitions in (a) for a field below 1 T. (c) Crossed-sections of σ^+ and σ^- spectra in (a) at 0.7 T. (d) “W” transition scheme at the field where FM and AFM levels anticross in (b). It is valid both for A^0-X^0 and A^0-X^- , hence only the selections rules (σ^+ or σ^-) and A^0 states are indicated.

a cross, can be found in Ref.’s [11, 19]. In brief, the FM and AFM lines correspond respectively to the ferromagnetic $|\pm 1, \pm 3/2\rangle$ and anti-ferromagnetic $|\pm 1, \mp 3/2\rangle$ spin configurations of A^0-X^- . They are split by the exchange energy $\Delta \sim 300\mu\text{eV}$, see Fig. 1(b), mostly due to

the A^0 - h ferromagnetic interaction while the A^0 - e^- exchange taking place in the final state of X^- optical recombination can be neglected [11]. A second very essential feature is the anisotropic coupling $\delta/2$ within the A^0 states $|\pm 1\rangle$. It gives rise to a level splitting $\delta \sim 50\mu\text{eV}$ at zero field both for A^0 or A^0 - e^- , and at finite magnetic fields $B = \pm\Delta/(2g_{A^0}\mu_B)$ for A^0 - X^0 or A^0 - X^- states when a FM and a AFM level are brought in coincidence by Zeeman effect (Fig. 1(b)). Here, g_{A^0} denotes the effective g -factor of A^0 spin $J = 1$ and μ_B is the Bohr magneton. For these specific fields we identify a “W” transition scheme (Fig. 1(d)) which, interestingly, reproduces the same scheme as used in Ref.’s [15, 16] to initialize and readout non-destructively an electron spin in a QD molecule. It is composed of a central Λ system allowing to induce A^0 spin flip transitions ($|+1\rangle \rightarrow |-1\rangle$ or $|-1\rangle \rightarrow |+1\rangle$) through σ^+ resonant excitation followed by spontaneous emission (*i.e.* OP), and two outer σ^- transitions allowing non-destructive readout of the A^0 spin as they do not change the A^0 state $|\pm 1\rangle$ while being cycled [20].

Let’s first examine more closely the A^0 - X^- PL spectrum under cw non-resonant and unpolarized excitation at the anticrossing field $B = 0.7$ T (Fig. 1(c)). The σ^- -resolved PL spectrum shows mainly two lines which correspond to the W outer transitions and therefore reflect the population probability $p_{\pm 1}$ of the $|\pm 1\rangle$ states. The low energy state $|-1\rangle$ is indeed found more populated than the $|+1\rangle$ state, yielding a spin polarization $(p_{-1} - p_{+1})/(p_{-1} + p_{+1})$ of $\sim 20\%$. However, the associated effective temperature determined from the PL intensity ratio amounts to ~ 10 K which is substantially higher than the lattice temperature [21]. This effective heating is partially due to the energy relaxation of the photo-carriers under non-resonant excitation. Yet, another key contribution comes from the optical recombination of σ^+ polarized trions through the Λ transitions which tend to equilibrate the $|+1\rangle$ and $|-1\rangle$ populations. This mechanism is evidenced by the σ^+ -resolved PL spectrum in Fig. 1(c) which shows two doublets of roughly equivalent intensity corresponding to both Λ branches. Eventually, σ^+ and σ^- PL spectra under non-resonant excitation are both encouraging indications to achieve OP and A^0 spin readout using this W system.

Obviously, non energy-selective excitation of the QD cannot lead to OP. Even with a polarized excitation creating only σ^+ -polarized trions the $|-1\rangle \rightarrow |+1\rangle$ conversion via the Λ transitions would be as probable as the $|+1\rangle \rightarrow |-1\rangle$ conversion. To favor one of them, an energy selective excitation is necessary, but in that case measuring the PL signal at almost the same energy turns out somewhat problematic. To circumvent this issue we

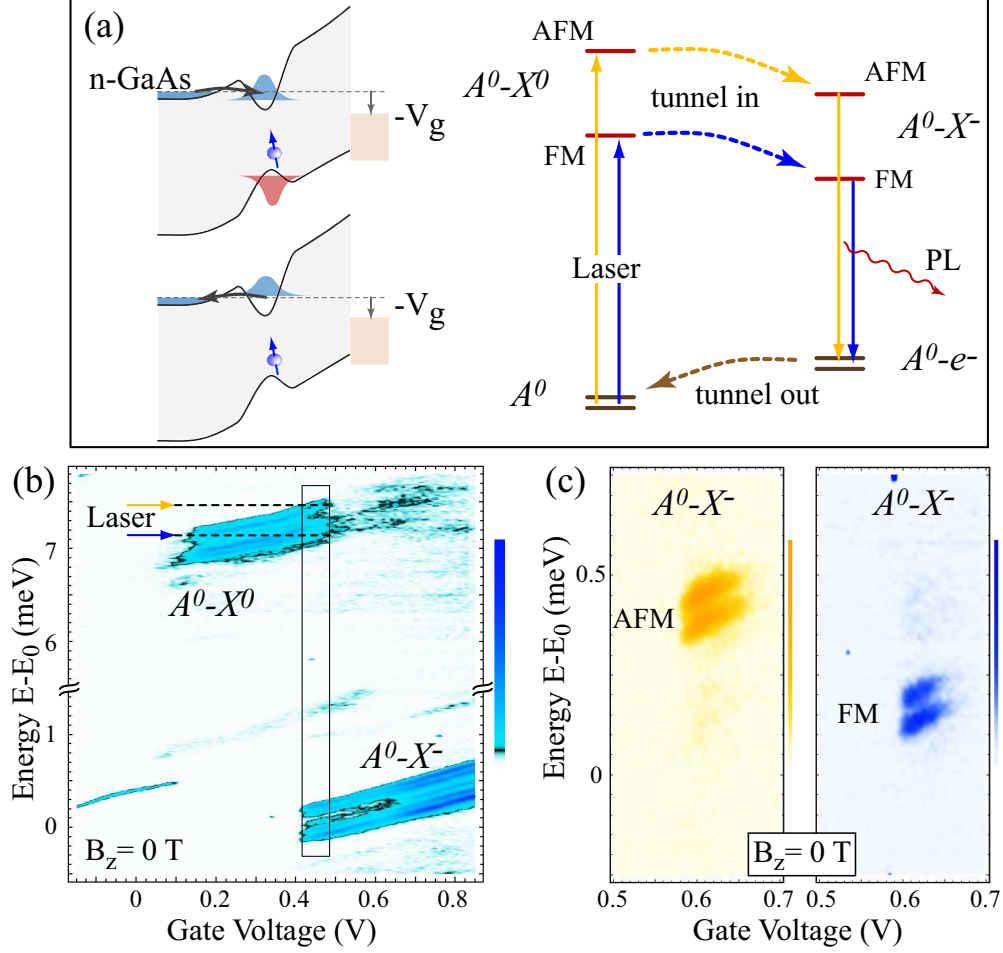


FIG. 2. (a) Schematics of the charge tuneable structure and of the X^0 - X^- cycle used for selective resonant excitation of a specific A^0 - X^0 spin configuration ($B_z = 0$ T). (b) PL intensity density plot of the Mn-doped QD against detection energy and gate voltage. The excitation is quasi-resonant around $E_0 + 50$ meV. (c) Similar density plot of X^- PL observed through the resonant excitation of either the FM or AFM X^0 level in a narrow voltage range around the X^0 - X^- frontier.

used a solution based on a singular excitation-emission cycle that takes place in a narrow range of applied gate voltages V_g at the frontier between X^0 and X^- [17, 22]. In this range, a resonantly created exciton X^0 is energetically unstable. It attracts in a time $\tau_c \sim 50$ ps much shorter than the radiative lifetime an electron which tunnels in the QD from the n-GaAs reservoir. This leads to the formation of an X^- trion which recombines optically producing a PL signal red-shifted by ~ 7 meV with respect to the excitation energy, after which the e^- left in the QD escapes back to the reservoir as depicted in Fig. 2(a). This cycle allows to resonantly excite X^0 with a laser energy outside of the spectrometer field of

view and to collect the red-shifted X^- PL signal in order to probe the A^0 spin. In practice, the internal diffusion of the laser inside the spectrometer can still blind the detector. Using crossed polarizers (H/V or σ^+/σ^-) between excitation and detection is necessary to reduce this stray light by a factor of a few 10^3 , while the remaining average background can be easily removed by taking a reference spectrum (with no PL signal) at a lower bias. The range of applied gate voltages for which this cycle occurs can be estimated by looking for the coexistence of X^0 and X^- spectral lines under quasi-resonant excitation (Fig. 2(b)). However, a significant voltage offset (up to +0.2 V) has to be applied when the excitation goes to resonance, to account for the reduction of the photocarrier-induced electric field screening.

As shown in Fig. 2(c), by resonantly exciting the X^0 upper (lower) doublet around $V_g = 0.62$ V we are able to detect the PL signal of the corresponding X^- upper (lower) doublet. The good preservation around 70% of the selected FM or AFM configuration validates our approach for selectively addressing and reading out A^0 spin states via the X^0 - X^- cycle. The unwished “cross-talking” is due to the large broadening of X^0 resonance of about 100 μeV [23] rather than a hole or A^0 spin-flip associated to the electron tunnelling in the dot. In this regards we benefit from the fact that all exchange interactions are essentially longitudinal (Ising type) because of the heavy character of the QD and A^0 hole, and therefore work together to conserve a given spin orientation during the e^- capture process [12]. Similarly, when the electron tunnels out of the dot after X^- recombination, one can reasonably expect that the weak A^0 - e^- interaction ($< 30 \mu\text{eV}$) which is also mainly longitudinal, will not imply any transitions between $|+1\rangle$ and $|-1\rangle$ states. In practice, the resonant excitation of X^0 appears to be equivalent to that of X^- with respect to A^0 spin, as it could be anticipated from their very similar PL spectra against magnetic field [11].

Thanks to its broad spectral resonance the X^0 - X^- cycle can be driven with a laser line at a fixed wavelength while sweeping the magnetic field from 0 to ~ 1.5 T. Figures 3(b),(c) show the resulting PL density plots for a linearly polarized laser line respectively centered on the upper (AFM) or lower (FM) doublet of A^0 - X^0 in zero field. As discussed above, the detection is linearly polarized orthogonally to that of the excitation. For a direct comparison, Fig. 3(a) displays a zoom of Fig.1(a) where both σ^+ (solid lines) and σ^- (dashed lines) measurements are reported. Around 0.7 T the W configuration aimed for OP arises and yields indeed drastic changes of the detected PL spectra : the Λ σ^+ lines corresponding

to the pumped level and exhibiting the anticrossing at 0.7 T almost completely vanish, while the outer σ^- lines corresponding to the populated A^0 spin state are conserved or even reinforced. As sketched in the insets of Fig. 3(b),(c), the laser energy determines which transition of the Λ is selected and in turn which A^0 spin state is pumped. Exciting the Λ upper or lower branch transfers the A^0 spin in the $|+1\rangle$ state (Fig. 3(b)) or $|-1\rangle$ state (Fig. 3(c)). Quantitatively, from the PL intensity ratio of the σ^+ and σ^- lines one can estimate the A^0 spin polarization achieved in both cases to about -55% and +60% respectively. This analysis relies on the assumption that the PL line intensities correctly reflect the initial $|+1\rangle$ and $|-1\rangle$ populations, whereas the laser is slightly blue- or red-detuned from the respective resonances. Such assumption sounds yet quite reasonable because the laser line was centered between the σ^+ and σ^- transitions.

To better evidence the effect of OP by using both outer branches of the W, as done to estimate the thermal polarization in Fig. 1(c), we performed a different experiment (Fig. 3(d)) : the resonant pump laser is now σ^+ circularly polarized and drives only the Λ upper branch, while a second quasi-resonant laser (at $E_0 + \sim 50$ meV) is used for the readout. Note that suppressing the pump laser stray light requires now a σ^- detection which in turn implies to use such quasi-resonant excitation for the σ^- probe laser. The main drawback of this approach is that the σ^- polarization of the photo-created excitons or trions is partially lost during the relaxation to the QD S-shell. This leads to some feeding of the Λ upper state, as illustrated in the inset of Fig. 3(d), which tends to depolarize the A^0 spin. Nevertheless, when turning on the pump laser, we clearly observe OP signature through the drastic increase (decrease) of the $|+1\rangle$ ($|-1\rangle$) line intensity. Under these non-optimal conditions a spin polarization, clearly opposite to the thermal one, of -49% is still reached. Conversely, by pumping the Λ lower branch (not shown) we could increase the thermal polarization from +21% to +55%. The imperfect excitation selectivity associated to the X^0 - X^- cycle is likely the main limitation of the uncomplete spin preparation achieved here. Truly resonant techniques should be employed to estimate properly the role of intrinsic A^0 spin relaxation.

In this Letter we have presented the efficient optical pumping and non-destructive readout of a single magnetic spin forming a 2-level system and embedded in an InAs/GaAs QD. Our results are stimulating for study further its potential as a qubit in condensed matter. In particular, thanks to the QD-induced anisotropy which provides this system with a nano-magnet character, one may anticipate a long spin relaxation time.

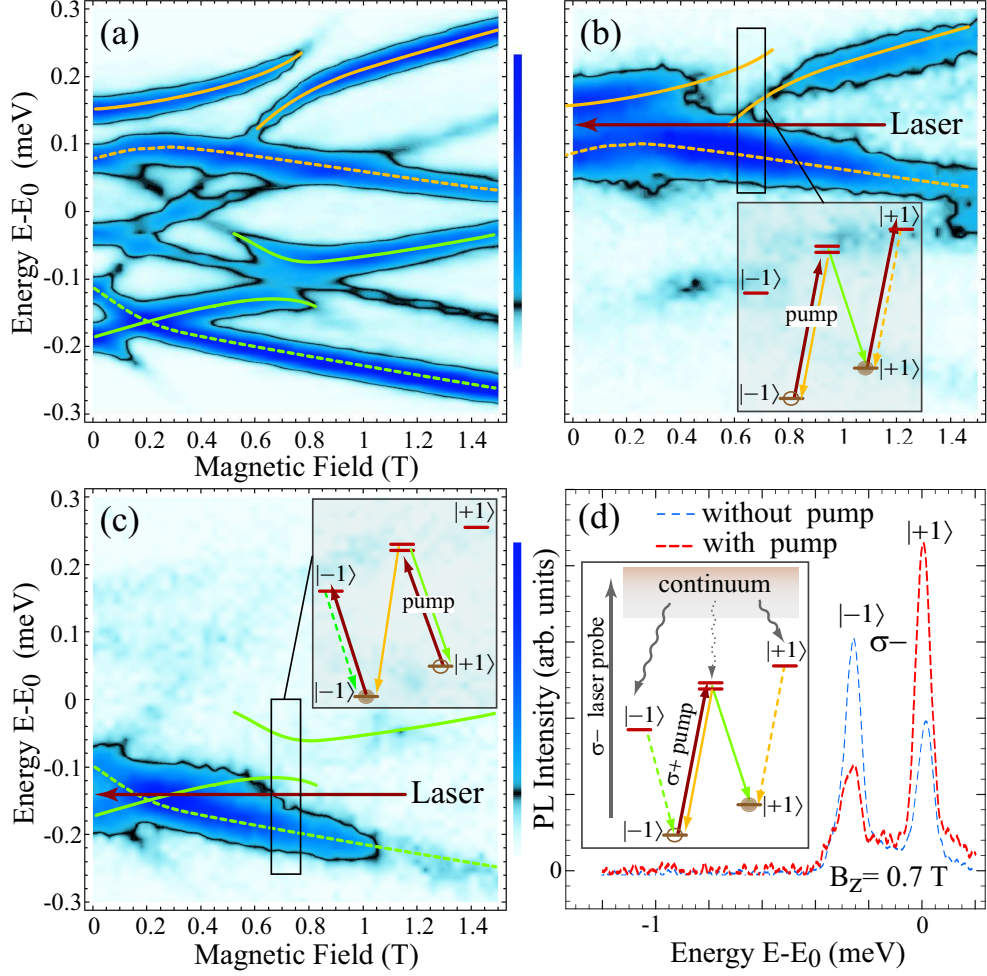


FIG. 3. (a) Zoom of Fig. 1(a) showing both σ^+ (solid lines) and σ^- (dashed lines) spectra. (b) and (c) PL density plots under resonant excitation centered on the zero field upper or lower A^0-X^0 doublet. The vanishing of the σ^+ anticrossing line around 0.7 T while the σ^- line persists or even increases indicates an efficient OP mechanism as depicted in the respective insets. (d) σ^- PL spectra at 0.7 T under σ^- quasi-resonant excitation (at $E_0 + \sim 50$ meV), with or without a resonant σ^+ excitation driving the upper Λ branch. The enhancement (reduction) of the $|+1\rangle$ ($|-1\rangle$) line with the pump laser demonstrates an efficient OP of -49%.

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